

Rapid diffusion of electrons in GaMnAs

C. P. Weber,^{1, a)} Eric A. Kittlaus,¹ Kassandra B. Mattia,¹ Christopher J. Waight,¹ J. Hagmann,² X. Liu,² M. Dobrowolska,² and J. K. Furdyna²

¹⁾Department of Physics, Santa Clara University, 500 El Camino Real
Santa Clara, CA 95053-0315, USA

²⁾Department of Physics, University of Notre Dame
Notre Dame, IN 46556, USA

(Dated: 4 March 2013)

We report ultrafast transient-grating measurements, in the paramagnetic state, of the dilute ferromagnetic semiconductor (Ga,Mn)As containing 6% Mn. We observe that photoexcited electrons in the conduction band have a lifetime of 8 ps and diffuse at about 70 cm²/s. Such rapid diffusion requires either an electronic mobility exceeding 7,700 cm²/Vs or a conduction-band effective mass m_e less than half the GaAs value. We further find that m_e cannot greatly exceed the GaAs value, as a large mass would require a long momentum-relaxation time. Our data suggest that neither the scattering rate nor the effective mass of the (Ga,Mn)As conduction band differs significantly from that of GaAs.

The following article is submitted to *Applied Physics Letters*.

The dilute magnetic semiconductor (Ga,Mn)As is considered an archetypal spintronic material. Despite years of intensive study, the band structure of its holes continues to excite debate.¹ The conduction band, on the other hand, is nearly unstudied. Its transport properties, however, bear importantly on the proposed “magnetic bipolar transistor,”^{2–4} which would operate through the diffusion current of electrons in a (Ga,Mn)As base. The device’s current gain and spin injection would both increase with increasing minority-carrier diffusivity.

We are not aware of measurements of electron transport in (Ga,Mn)As, and the extensive study of the valence band offers only mixed guidance as to what might be expected of the conduction band. Recent experiments indicated a long scattering time⁵ and GaAs-like effective mass^{5,6} in the valence band. On the other hand, the hole mobility of (Ga,Mn)As is typically low,⁷ likely due to disorder resulting from Mn doping, and the mean free path is estimated as just a few nanometers.⁸ The Mn-induced disorder has been imaged by scanning tunneling microscopy, and persists from the valence band through the conduction-band edge,⁹ which suggests a short scattering time for electrons.

Here we measure that diffusion of photoexcited electrons, during the first several picoseconds after excitation, is about 70 cm²/s. Such high diffusivity implies that the electrons’ mobility must be high, or their effective mass low, or both. In particular, our data are consistent with an electron effective mass equal to that of GaAs and a mobility greater than 7,700 cm²/Vs.

Our (Ga,Mn)As sample was grown by molecular beam epitaxy on a substrate of (001) semi-insulating GaAs. The structure consists of a 250 nm stop-etch layer of Al_{0.35}Ga_{0.65}As, a 25 nm buffer of GaAs, and the 800 nm sample of Ga_{0.94}Mn_{0.06}As, grown with a Mn cell temper-

ature of 760 °C and a substrate temperature of 215 °C. After growth the film was affixed to a sapphire window and the GaAs substrate removed by chemical etching.¹⁰ Since Al_{0.35}Ga_{0.65}As is transparent at our measurement wavelength of 810 nm, our signal arises entirely in the (Ga,Mn)As film. The film’s resistivity indicates a Curie temperature $T_C \approx 56$ K. From these data we estimate the hole density as $p_0 \approx 1 \times 10^{20}$ cm⁻³. The sample has a resistivity $\rho = 12$ mΩcm at 80 K, the temperature of this experiment.

We measured diffusion by the ultrafast transient-grating method, which measures the lifetime τ of a density wave (“grating”) of photoexcited electrons and excess holes. The grating’s wavelength is Λ , its wavevector $q = 2\pi/\Lambda$, and its amplitude decays at a rate of

$$\frac{1}{\tau(q)} = D_a q^2 + \frac{1}{\tau_0}. \quad (1)$$

D_a is the ambipolar diffusion coefficient and τ_0 is the lifetime of spatially uniform excitation. Measurement at several q determines D_a . We measure the diffracted probe amplitude in a reflection geometry, improve the efficiency by heterodyne detection,¹¹ and suppress noise by 95-Hz modulation of the grating phase and lock-in detection.¹²

The laser pulses have wavelength near 810 nm, duration 120 fs, and repetition rate 80 MHz. The pump pulses, whose interference and absorption creates the grating, have fluence at the sample of 4.2 μJ/cm²; the probe pulses are a factor of 12 weaker. At 810 nm GaAs has an absorption length¹³ of order 1 μm and reflectivity of 0.3, so at our highest fluence each pair of pump pulses photoexcites electrons and holes at a mean density of $n_{\text{ex}} \approx 1.2 \times 10^{17}$ cm⁻³. The incident photons have energy about 30 meV greater than the bandgap of GaAs at 80K. (Ga,Mn)As is known to suffer bandgap narrowing of order 120 meV,¹⁴ so we estimate that our photoexcited electrons have excess energy of 150 meV. Screening of the electron-LO phonon interaction¹⁵ could slow the

^{a)}Electronic mail: cweber@scu.edu

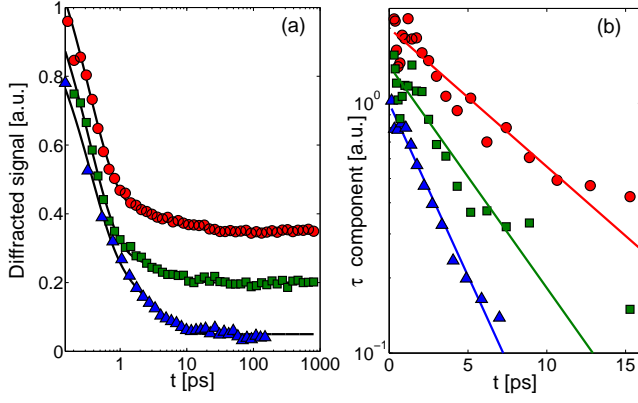


FIG. 1. (Color online) Diffracted amplitude $S(t)$ for $q = 12,600 \text{ cm}^{-1}$ (circles), $37,700 \text{ cm}^{-1}$ (squares), and $52,800 \text{ cm}^{-1}$ (triangles). Scaled and offset vertically for clarity. Solid lines are fits to the form of Eq. 2. (a) Entire signal; semilog t . (b) Signal with constant and τ_{fast} terms subtracted, leaving just the $\tau(q)$ component; semilog S .

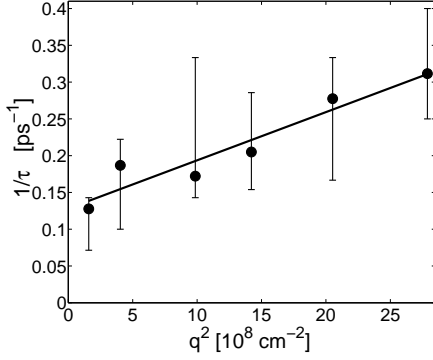


FIG. 2. Decay rate of the signal's $\tau(q)$ component *vs.* q . Points and error bars are obtained by procedures described in the text. The line is a least-squares fit of the points to Eq. 1, revealing diffusion of photoexcited carriers.

electrons' cooling to the lattice temperature, but they will rapidly thermalize with the much more numerous holes through carrier-carrier scattering. The holes' specific heat is about 100 times greater than the electrons', resulting in a total heating of just 20 K. Thus the rapid diffusion that we observe does not reflect the transport properties of hot electrons, but of equilibrium ones.

Diffusivity varies¹⁶ with n_{ex} , and the peaks of our grating will diffuse more rapidly than the troughs. Our numerical simulations indicate that for the densities and temperature used in this experiment, this effect can cause us to underestimate D_a by up to 30%, making electron mobility potentially somewhat higher than the value we report.

Figure 1a shows the diffracted signal $S(t)$ as a function of time for gratings of several q . In each case the signal is well described by the form

$$S(t) = Ae^{-t/\tau_{\text{fast}}} + Be^{-t/\tau(q)} + C, \quad (2)$$

with $\tau_{\text{fast}} \approx 0.34 \text{ ps}$, $3 \text{ ps} < \tau(q) < 8 \text{ ps}$, $B/A \approx 0.1$, and $C/A \approx -0.015$. Of these three components of our signal, we will see below that the $\tau(q)$ term reveals the dynamics of free electrons diffusing at about $70 \text{ cm}^2/\text{s}$.

Previous ultrafast experiments on (Ga,Mn)As have observed signals with a very slow component corresponding to our C term, and attributed it to trapped carriers^{17–19} or to residual heat from the excitation pulse.^{20,21} Such experiments have also observed fast processes on the scale of τ_{fast} ,^{17–19} which were variously attributed to intraband relaxation¹⁸ or trapping¹⁷ of electrons at As antisite defects. Rapid trapping is not inconsistent with our observation of electrons' diffusion at later times, since the number of electrons photoexcited likely exceeds the number of trapping sites. Our observations do, however, contradict reports²⁰ that all carriers are trapped and recombined within a picosecond.

Since D_a is determined from $\tau(q)$, it is critical to find the latter values reliably, despite the constant and τ_{fast} terms, which introduce free parameters when fitting the data to Eq. 2. By measuring $S(t)$ at times much greater than $\tau(q)$, we determine C unambiguously. Since τ_{fast} appears to have similar values at all q , we reduce parameters further by fitting all the data with the single value $\tau_{\text{fast}} = 0.34 \text{ ps}$. The resulting values of $1/\tau(q)$ appear as the points in Fig. 2. That this procedure gives reliable fits to the data's $\tau(q)$ component is evident from Fig. 1b, in which we plot the measured $S(t)$ with the constant and τ_{fast} terms subtracted. The signal decays exponentially, and the decay is visibly faster at high q , indicative of diffusive motion. Finally, we repeat each fit for a series of *fixed* values of $\tau(q)$ and judge “by eye” for what range of $\tau(q)$ the fits appear to plausibly reproduce the data. The outer limits of this range determine the error bars shown in Fig. 2.

The line in Fig. 2 is a fit to the form of Eq. 1, giving $D_a = 67 \text{ cm}^2/\text{s}$ and $\tau_0 = 8 \text{ ps}$. (Fits to the lower or upper ends of the error bars give values of D_a ranging from 60 to $80 \text{ cm}^2/\text{s}$, and τ_0 from 14 to 6 ps.) This figure makes it evident that our signal's τ -component arises from ambipolar diffusion of free electrons, for the diffusivity is much faster than expected for heat or trapped carriers.

Though 8 ps may seem too short to be the free-electron lifetime, it is consistent with previous observations in low-temperature-grown GaAs^{22,23} and in (Ga,Mn)As,^{18,24–27} where a transient lasting 6 to 14 ps was attributed to non-radiative recombination of trapped carriers.^{25,26} Our signal arises from free electrons, not trapped, but it may be sensitive to recombination because each recombination event empties a trap, allowing another free electron to be trapped. In what follows, n_f , the number of *untrapped* electrons, will be important. Our “best guess” is that $n_f/n_{\text{ex}} \approx B/A \approx 0.1$, *i.e.*, that the relative amplitude of the $\tau(q)$ and τ_{fast} terms represents the proportion of electrons remaining untrapped after 0.34 ps. Note, however, that our conclusions will hold true for the entire possible range $0 < n_f < n_{\text{ex}}$.

We now show that it is possible to use our measure-

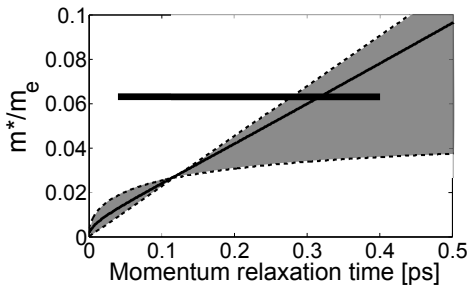


FIG. 3. For the simple model described, the relation between the electrons' momentum relaxation time t_p and their effective mass m^*/m_e , as constrained by our data. The solid curve assumes a density of untrapped carriers $n_f = n_{\text{ex}}/10$. The shaded region is bounded by $n_f = n_{\text{ex}}$ (curved) and $n_f = 0$ (diagonal). The mobility exceeds $7,700 \text{ cm}^2/\text{Vs}$ for all points below the diagonal boundary. The thick line is the range of values expected for GaAs at 80 K.

ments of D_a and resistivity ρ (of the holes) to infer conduction-band properties, despite our uncertain knowledge of three key parameters. The first is μ_e , the electrons' mobility; second is n_f . Finally, the electrons' charge-susceptibility is $\chi_e = dn/d\mu_{\text{chem}}$, with μ_{chem} the chemical potential. This quantity approaches $n_f/k_B T$ at high temperature, and at low temperature approaches N_e , the electrons' density of states at their quasi-Fermi energy; it thus provides insight into the electrons' effective mass m^* . (With χ_h defined similarly for holes, the reduced susceptibility is $1/\chi^* \equiv 1/\chi_e + 1/\chi_h$. In this experiment $\chi_h > \chi_e$ and $\chi^* \approx \chi_e$; we nonetheless include the hole susceptibility in our numerical calculations.²⁸)

By the Einstein relation, the electrons' diffusivity is

$$D_e = \frac{n_f \mu_e}{e \chi_e}, \quad (3)$$

and similarly for holes. The photoexcited electrons and holes must diffuse together, giving ambipolar diffusion²⁹ $D_a = (\sigma_e D_h + \sigma_h D_e)/(\sigma_e + \sigma_h)$, from which

$$\frac{1}{D_a} = \left(e^2 \rho + \frac{e}{n_f \mu_e} \right) \chi^*. \quad (4)$$

Here we have used $n_{\text{ex}} \ll p_0$. Our measurements suffice to specify neither χ^* nor μ_e , but do constrain the relation between them: Eq. 4 shows that the larger the mobility, the larger χ^* .

To further explore the relationship between μ_e and χ^* , it will be instructive to recast it in terms of m^* and the momentum relaxation time t_p , and to compare our results on (Ga,Mn)As with the known properties of GaAs (which has $m^*/m_e = 0.063$). In undoped GaAs the mobility at 80 K can reach $2 \times 10^5 \text{ cm}^2/\text{Vs}$, but for $n \approx n_{\text{ex}}$ the mobility should be 10^3 to $10^4 \text{ cm}^2/\text{Vs}$.^{30,31} The latter values correspond to momentum relaxation times t_p of 0.04 to 0.4 ps. The range of typical GaAs parameters appears as a thick, horizontal line in Fig. 3, where we choose to plot t_p rather than μ_e since mobility contains both t_p and m^* .

Turning back to our (Ga,Mn)As results, we note that at finite temperature $\chi^*(m^*)$ must be calculated numerically.³² For this purpose we introduce a simple model that treats both bands as isotropic and parabolic, the hole mass²⁸ as $0.53m_e$, and the conduction-band effective mass m^* as a free parameter. We calculate first $\chi^*(m^*)$, then the corresponding $\mu_e(\chi^*, D_a, \rho)$, and finally the momentum relaxation time $t_p = \mu_e m^*/e$; the results appear in Fig. 3. Since n_f is unknown, we repeat the calculation for a range of n_f : the solid curve assumes $n_f/n_{\text{ex}} = 0.1$, and we shade the entire possible region $0 < n_f/n_{\text{ex}} < 1$. Note that all points on the line for $n_f = 0$ have $\mu_e = 7,700 \text{ cm}^2/\text{Vs}$, and that points in the allowed region below the line have yet higher mobility. We believe, therefore, that $\mu_e \geq 7,700 \text{ cm}^2/\text{Vs}$; this value is high, but not without precedent in GaAs.

It may seem that (Ga,Mn)As, as a disordered material, ought to have mobility well below that of GaAs. Indeed, our experimental results do not, strictly, exclude a low mobility: it is allowed if $m^*/m_e < 0.03$. We are not, however, aware of any mechanism to suppress the conduction-band density of states so strongly. It would also seem unlikely for t_p to much exceed 0.4 ps, which suggests that m^* cannot much exceed the GaAs value. Thus the most plausible scenario supported by our data is that neither the conduction band's scattering rate nor its effective mass is much influenced by Mn doping. We note that some recent results indicated that the (Ga,Mn)As valence band has GaAs-like effective mass^{5,6} and a low scattering rate⁵. Indeed, those results are even more surprising than ours, because Mn states are much closer to the valence band edge than to the conduction band.

The high diffusivity we measure could aid the operation of a GaMnAs-based magnetic bipolar transistor.²⁻⁴ In order to exhibit high current gain, the device would need a minority-carrier diffusion length that greatly exceeds the base width w . The electrons' diffusion length of $L = \sqrt{D_e \tau_0} \approx 240 \text{ nm}$ is not long, but is long enough to allow $w < L/10$ to be achievable by commercially-available photolithography techniques. Such a device would inject a spin polarization proportional to D_e into the collector.³³

In conclusion, we performed transient-grating experiments at 80 K to measure the diffusion of photoexcited electrons in $\text{Ga}_{0.94}\text{Mn}_{0.06}\text{As}$. We found a signal consisting of three components; the component revealing motion of free electrons had a lifetime of 6-14 ps and a diffusivity of $60\text{-}80 \text{ cm}^2/\text{s}$. This rather high diffusivity, along with the measured resistivity, constrains the electrons to have either a high mobility or very a low effective mass. We consider it most plausible that the electrons' effective mass and mobility are both comparable to those of GaAs, with $\mu_e \geq 7,700 \text{ cm}^2/\text{Vs}$. The mobility is nonetheless surprisingly high, given the disorder in (Ga,Mn)As alloys, and reflects favorably on the possibility of GaMnAs-based magnetic bipolar transistors. Our result suggests the value of further measurements, such as a Shockley-Haynes experiment, that would measure μ_e directly and

would thus allow the determination of both the electrons' mobility and their effective mass.

This work was supported by the National Science Foundation Grants No. DMR-1105553 and DMR 10-05851. E.A.K. and K.B.M. were partly supported by Santa Clara University's Hayes and Clare Booth Luce Scholarships, respectively.

- ¹N. Samarth, *Nature Mater.* **11**(5), 360 (2012).
- ²J. Fabian, I. Žutić, and S. Das Sarma, *Phys. Rev. B* **66**(16), 165301 (2002).
- ³M. E. Flatte, Z. G. Yu, E. Johnston-Halperin, and D. D. Awschalom, *Appl. Phys. Lett.* **82**(26), 4740 (2003).
- ⁴N. Lebedeva and P. Kuivalainen, *J. Appl. Phys.* **93**(12), 9845 (2003).
- ⁵S. Ohya, K. Takata, and M. Tanaka, *Nature Phys.* **7**(4), 342 (2011).
- ⁶B. C. Chapler, S. Mack, L. Ju, T. W. Elson, B. W. Boudouris, E. Nanddas, J. D. Yuen, A. J. Heeger, N. Samarth, M. Di Ventra, *et al.*, *Phys. Rev. B* **86**(16), 165302 (2012).
- ⁷T. Jungwirth, J. Sinova, A. H. MacDonald, B. L. Gallagher, V. Novak, K. W. Edmonds, A. W. Rushforth, R. P. Campion, C. T. Foxon, L. Eaves, *et al.*, *Phys. Rev. B* **76**(12), 125206 (2007).
- ⁸C. P. Moca, B. L. Sheu, N. Samarth, P. Schiffer, B. Janko, and G. Zarand, *Phys. Rev. Lett.* **102**(13), 137203 (2009).
- ⁹A. Richardella, P. Roushan, S. Mack, B. Zhou, D. A. Huse, D. D. Awschalom, and A. Yazdani, *Science* **327**(5966), 665 (2010).
- ¹⁰J. H. Kim, D. H. Lim, and G. M. Yang, *J. Vac. Sci. & Technol. B* **16**(2), 558 (1998).
- ¹¹P. Vohringer and N. F. Scherer, *J. Phys. Chem.* **99**(9), 2684 (1995).
- ¹²C. P. Weber, N. Gedik, J. E. Moore, J. Orenstein, J. Stephens, and D. D. Awschalom, *Nature* **437**(7063), 1330 (2005).
- ¹³M. D. Sturge, *Phys. Rev.* **127**(3), 768 (1962).
- ¹⁴Y. Ohno, I. Arata, F. Matsukura, and H. Ohno, *Physica E* **13**(24), 521.
- ¹⁵S. Das Sarma, A. Kobayashi, and W. Y. Lai, *Phys. Rev. B* **36**(15), 8151 (1987).
- ¹⁶C. P. Weber and E. A. Kittlaus, *J. Appl. Phys.* **113**(5), 053711 (2013).
- ¹⁷J. P. Zahn, A. Gamouras, S. March, X. Liu, J. K. Furdyna, and K. C. Hall, *J. Appl. Phys.* **107**(3), 033908 (2010).
- ¹⁸S. Kim, E. Oh, J. U. Lee, D. S. Kim, S. Lee, and J. K. Furdyna, *Phys. Status Solidi C* **2**(8), 3141 (2005).
- ¹⁹S. Kim, E. Oh, J. U. Lee, D. S. Kim, S. Lee, and J. K. Furdyna, *Appl. Phys. Lett.* **88**(26), 262101 (2006).
- ²⁰E. Kojima, R. Shimano, Y. Hashimoto, S. Katsumoto, Y. Iye, and M. Kuwata-Gonokami, *Phys. Rev. B* **68**(19), 193203 (2003).
- ²¹E. Kojima, J. B. Heroux, R. Shimano, Y. Hashimoto, S. Katsumoto, Y. Iye, and M. Kuwata-Gonokami, *Phys. Rev. B* **76**(19), 195323 (2007).
- ²²H. S. Loka, S. D. Benjamin, and P. W. E. Smith, *IEEE J. Quantum Electron.* **34**(8), 1426 (1998).
- ²³M. Stellmacher, J. Nagle, J. F. Lampin, P. Santoro, J. Vaneecloo, and A. Alexandrou, *J. Appl. Phys.* **88**(10), 6026 (2000).
- ²⁴Y. Mitsumori, A. Oiwa, T. Slupinski, H. Maruki, Y. Kashimura, F. Minami, and H. Munekata, *Phys. Rev. B* **69**(3), 033203 (2004).
- ²⁵B. Q. Sun, D. S. Jiang, Z. Sun, X. Z. Ruan, J. J. Deng, J. H. Zhao, Y. Ji, and Z. Y. Xu, *J. Appl. Phys.* **100**(8), 83104 (2006).
- ²⁶E. Rozkotova, P. Nemec, P. Horodyska, D. Sprinzl, F. Trojanek, P. Maly, V. Novak, K. Olejnik, M. Cukr, and T. Jungwirth, *Appl. Phys. Lett.* **92**(12), 122507 (2008).
- ²⁷X. D. Liu, W. Z. Wang, R. X. Gao, J. H. Zhao, J. H. Wen, W. Z. Lin, and T. S. Lai, *Acta Phys. Sin.* **57**(6), 3857 (2008).
- ²⁸ χ_h depends on p_0 and the hole density of states, neither of which is accurately known. However, the effect of the holes on χ^* is less than 8% provided p_0 exceeds 10^{18} cm^{-3} and the hole effective mass exceeds 1/20 of the free-electron mass.
- ²⁹W. van Roosbroeck, *Phys. Rev.* **91**(2), 282 (1953).
- ³⁰Rode, D. L., in *Semiconductors and Semimetals*, Vol. 10: Transport Phenomena, edited by R. K. Willardson and A. C. Beer (Academic Press, 1975).
- ³¹J. R. Lowney and H. S. Bennett, *J. Appl. Phys.* **69**(10), 7102 (1991).
- ³²N. Mohankumar and A. Natarajan, *Phys. Status Solidi B* **188**(2), 635 (1995).
- ³³J. Fabian and I. Žutić, *Phys. Rev. B* **69**(11), 115314 (2004).